Mass Spectrometry Imaging of molecular biomarkers on intact sediment sections

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Molecular biomarkers are organic compounds diagnostic for a specific organism, community or biological process that can be employed to trace them in present or past ecosystems. Some compounds, or compound ratios, are further found to correlate with a certain physical property, and can therefore serve as a proxy for reconstruction of temporal changes of this variable. When being produced in the water column, after sedimentation – and due to geochemical persistence in the sedimentary record – the information archived in molecular biomarkers is preserved over geological time-scales and serves as an important informant of past environments. Conventional biomarker analysis is labor-intensive and requires large samples that limit temporal resolution. We present a Mass Spectrometry Imaging (MSI) approach, based on laser desorption ionization coupled to ultra high resolution mass spectrometry, that avoids wet-chemical sample preparation and enables spatially resolved analysis directly on sediment sections at sub-mm spatial resolution.

Our initial study targeted archaeal glycerol dialkyl glycerol tetraethers (GDGTs) that are ubiquitous in marine sediments and serve as valuable paleoenvironmental proxies. GDGT-profiles yielded paleoecologic and paleoenvironmental information at unprecedented temporal resolution and revealed a first glimpse of what appears to be fine-scale patchiness of biomarker distribution. To further strengthen these two aspects, additional developments are underway and a dedicated facility has been set up at MARUM/University of Bremen. Sample preparation for MSI has been optimized. Methods targeting other lipid biomarkers (e.g., alkenones or sterols) are being implemented and applied to recent laminated sediments (Santa Barbara Basin, Gotland Basin), yielding informative profiles with subannual resolution. And the combination of biomarker MSI with other spatially resolved techniques (e.g. elemental mapping by microXRF) is being employed to explain small scale variability and to provide a better understanding of biomarker distribution and signal formation.